Stress compensating multilayers

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ABSTRACT

We present in-situ stress measurement results for single and multilayer thin-films deposited by magnetron sputtering. In particular, we report on the influence of the material interfaces on the ensuing stress in both the transient and steady-state regimes of film growth. This behavior is used to determine the appropriate thicknesses of the constituent layers that will result in a net tensile stress in multilayers composed of various material combinations. These multilayers can then be used to compensate the compressive integrated stress in single and multilayer EUV and x-ray optical coatings. The use of multilayers to compensate the integrated stress might be advantageous because, unlike single layers of chromium, the roughness is not expected to increase with the total thickness of the multilayer. In this paper, we demonstrate the technique for W/Si and Mo/Si multilayers and discuss its application to other material combinations.

Keywords: In-situ stress measurement, stress compensation, x-ray optics

1. INTRODUCTION

The stress in thin films is often a leading technological challenge in the development of high resolution x-ray and EUV imaging optics. This is particularly true for the coating development for space-borne x-ray observatories for both past missions like *NuStar,* as well as future missions such as *Lynx* and *Athena*. The mitigation of substrate deformation caused by high integrated film stress will also be an important consideration for the successful future development of multilayer coatings for high energy x-ray astronomy. To illicit a broadband response at higher photon energies will require smaller d-spacings and a greater number of layers than coatings used for previous missions like *NuStar*. So that even for material combinations that exhibit relatively low intrinsic stress, such as W/Si, the integrated stress will still be appreciable due to the micron-scale total thickness of the layer stack.

The use of a chromium (Cr) thin film in a state of tensile stress, and of an appropriate thickness, has demonstrated the capability to balance the high compressive integrated stress in films like iridium (Ir) to near-zero values. This technique, however; has limited applicability because the film's microstructure is columnar which results in a continued increase in surface roughness as the film grows in thickness. This is particularly problematic for compensating the stress in multilayer coatings. For example, the maximum high energy response of broadband depth graded multilayer structures will be determined by the smallest d-spacings in the layer stack. The minimum bi-layer thickness will in turn be determined by the ability to deposit these layers with smooth, abrupt material interfaces. Since the smallest d-spacing in these broadband structures are deposited adjacent to the substrate, their interfacial quality depends in part on the smoothness of the surface upon which they are deposited. Others have demonstrated the capability to deposit periodic multilayers with d-spacings in the range of 10-15Å, but good reflectivity relies on a substrate roughness of ~3Å or less, for example[1]. The thickness of the chromium layer that would be needed to compensate the stress in high energy broadband multilayer would result in a surface roughness that substantially exceeds this requirement (i.e. 7-8 Å).

In an attempt to obviate this shortfall, we propose a method here which utilizes a multilayer in tensile stress, rather than a single (Cr) layer, to compensate the compressive stress in x-ray optical coatings. The advantage of this approach is that the thickness of the metal layer, which drives the net tensile stress in the layer stack, can be made much thinner than what would otherwise be required using a single layer compensating film like Cr. Therefore, reasonably large values of integrated stress can be compensated, to near-zero values if desired, simply by increasing the number of layers in the stack without an ensuing increase in surface roughness. Eligible material pairs and the thicknesses of their constituent layers needed to achieve a net tensile stress in the multilayer while also minimizing deposition time are easily identified from in-situ stress measurement. In this paper we demonstrate how the stress can be compensated in W/Si and Mo/Si optical coatings using a stress compensating multilayer composed of the same materials. The use of W/Si was a material pair that was used for the *NuStar* coatings. It remains a material pair of interested in the continued development of broadband multilayers beyond ~68 keV. The use Mo/Si, on the other hand, has been a popular material pair choice in the development of multilayers for EUV projection lithography where the stress in the coating can also limit the

resolution capability of the optics. The intent here is to illustrate proof of principle; additional work is needed to understand some of the underlying mechanisms and to further optimize the thickness of the layers to minimize deposition time and surface roughness. Further work is also needed to understand how the substrate temperature or deposition rate might impose any restriction on the applicability of the method.

1.1 In-situ stress measurement methodology

As we will demonstrate, in-situ stress measurement capability is an invaluable tool for identifying techniques and process mechanism to control the integrated stress in x-ray optical coatings. In this paper, it is used to identify potential material pairs and their corresponding layer thicknesses for use as a stress compensating multilayer. Briefly, the in-situ stress measurement apparatus utilizes a clamped cantilever substrate. The displacement of its free end, δ, is measured using a vacuum compatible fiber optic displacement sensor. This displacement is proportional to the integrated stress, σh_f (stress x film thickness) according to the Stoney equation which, in the form appropriate for a cantilever-substrate is,

$$\sigma h_f = \frac{E_s h_s^2 \delta}{3(1-\nu_s)L^2} \tag{1}$$

,where L is the cantilever's length, h_s its thickness, E_s its Young's modulus, and ν_s its Poisson's ratio. From measurement of the normal distribution of random noise of the cantilever tip we have determined the minimum detectable integrated stress to be 9 MPa-nm for a 100 µm thick Schott D263 glass substrate. This means, for example, that a change in film thickness of ~ 0.9Å could be detected for a film with a stress of 100 MPa. A more detailed explanation of the apparatus and its full capability can be found elsewhere[2].

2. IN-SITU STRESS MEASUREMENT RESULTS

The single and multilayer films presented here have been deposited by magnetron sputtering at an argon pressure of 2.5 mTorr. The films were deposited on 150 µm thick Schott D263 glass substrates that were cut into the shape of 37.5 X 5.7 mm rectangular cantilevers. The substrates were grounded during all of the depositions. The films were deposited using 3 in. diameter circular magnetron sputtering source that is in a fixed position relative to the stationary substrate. The multilayers were produced by switching between cathodes using a programmable power supply with a 0.1 sec delay between the deposition of each layer.

2.1 The influence of the material interfaces

Insight into the behavior of the net stress in the multilayers can be obtained by measuring the in-situ stress in a simple tri-layer system composed of a dielectric layer "sandwiched" between two metal layers of the same material. This measurement scheme provides the essential information needed to reasonably approximate the steady-state behavior of the cumulative stress in the multilayer as a function of the thicknesses of the constituent layers. The influence of diffusion and/or alloying at the material interfaces is necessarily measured which includes the effect of the energy and the "wettability" of the surface. As we will show, the material interfaces can have a significant contribution to the behavior of the stress in the multilayer.

2.1.1 Mo/Si, Mo/B_4C, Cr/B_4C interfaces ("low" density metals)

Fig. 1 and 2 shows the cantilever tip displacement as a function of deposition time for Mo/Si and Mo/B_4C interfaces, respectively. The stress in the Mo layer is tensile in the steady-state regime when deposited on glass and remains when deposited on a layer of amorphous silicon (Si) or boron carbide (B_4C). A similar behavior is observed for chromium (Cr) which is shown in Fig. 3 for the case of a B_4C interface, and although not explicitly shown here, is also expected to remain tensile in the steady-state when deposited on a layer of sputtered (Si) as well. The tensile stress in these metals is thought to arise because of the low adatom mobility resulting from the low temperature (27-30^{o}C) of the substrate during the deposition process. The adatom mobility may be further impeded by the lower atomic mass of these metals which arrive at the substrate with less initial kinetic energy in comparison to more massive metal atoms like tungsten (W), for example. As we have shown previously for Cr, these low density metals will also exhibit compressive stress in the steady-state at low argon pressure provided the substrate temperature is high enough to provide the necessary energy to

the adatoms[3]. We further expect that the substrate temperature at which the stress changes sign, denoted as the transition temperature, will scale inversely with the mass of the sputtered atoms for a given process pressure. The effect of substrate temperature is an important consideration and can differ depending on the heat transfer mechanisms that are specific to a given deposition geometry, substrate emissivity and mounting scheme. The deposition rate of the sputtered material also influences the substrate temperature, and hence, the film stress. Here the deposition rate for the metal films is in the range of 0.1-0.2 nm/sec.

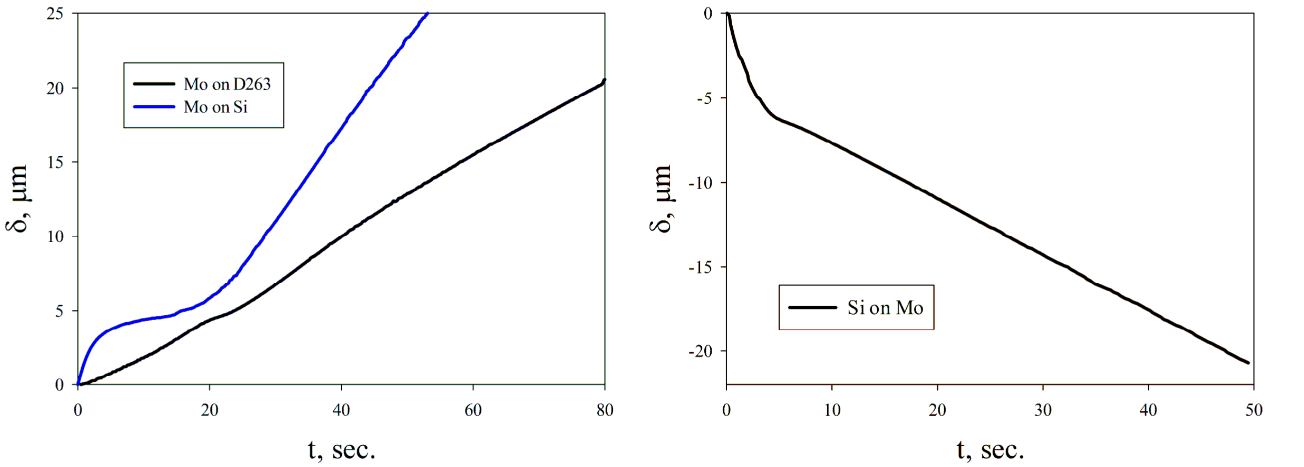

Figure 1. The cantilever tip displacement, δ, as a function of deposition time for the Mo on glass, Mo on Si (Mo/Si), and Si on Mo (Si/Mo) interfaces. The stress in the Mo layer remains tensile in the steady-state regime when deposited on a sputtered layer of silicon.

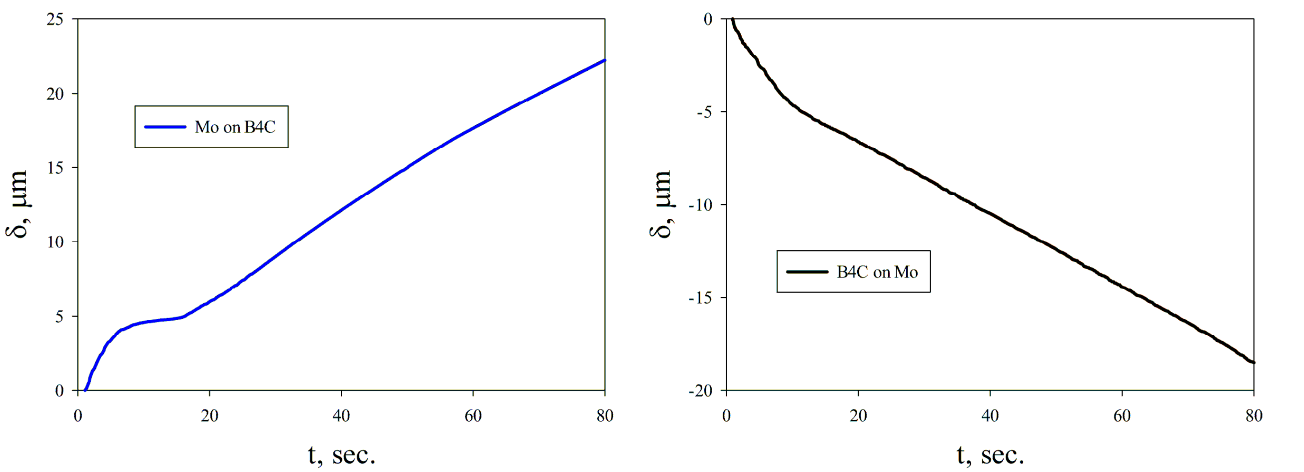

Figure 2. The cantilever tip displacement, δ, as a function of deposition time for the Mo on glass, Mo on B_4C (Mo/B_4C), and B_4C on Mo (B_4C/Mo) interfaces.

It seems that the low density metals that exhibit tensile stress in the steady-state regime when deposited at low argon pressure on glass, and at low enough substrate temperature, will continue to exhibit the same behavior when deposited on low-Z materials such as Si and B_4C. We therefore surmise that tensile stress can also be achieved for nickel (Ni) based multilayers as well because the density of Ni is somewhere between that of Mo and Cr; this will be verified in future work.

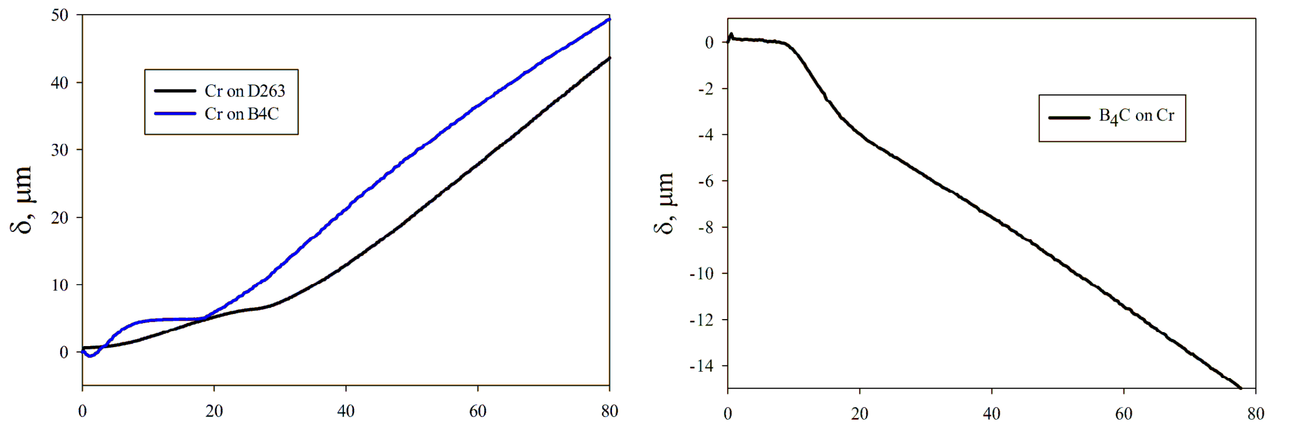

Figure 3. The cantilever tip displacement, δ, as a function of deposition time for the Cr on glass, Cr on B_4C (Cr/B_4C), and B_4C on Cr (B_4C/Cr) interfaces.

2.1.2 Ir/Si, W/Si interfaces ("high" density metals)

Fig. 4 shows the cantilever tip displacement as a function of deposition time for the W/Si material combination, along with the indicated material interface. The in-situ stress for the tungsten (W) single layer film deposited on glass exhibits the typical Volmer-Weber growth mode that is associated with high adatom mobility. In this growth mode, individual islands nucleate and then grow to eventually coalesce to form a continuous film. The tensile maximum indicates the moment at which island coalescence occurs, thereafter; the stress in growing film reaches a compressive steady-state. Here the deposition temperature is still low (27-30^{o}C), but it is thought that the tungsten atoms arriving at the surface of the growing film have sufficient kinetic energy, due to their higher mass in comparison to Cr and Mo, to provide high enough adatom mobility to promote compressive stress in the steady-state. Tungsten exhibits a different transient

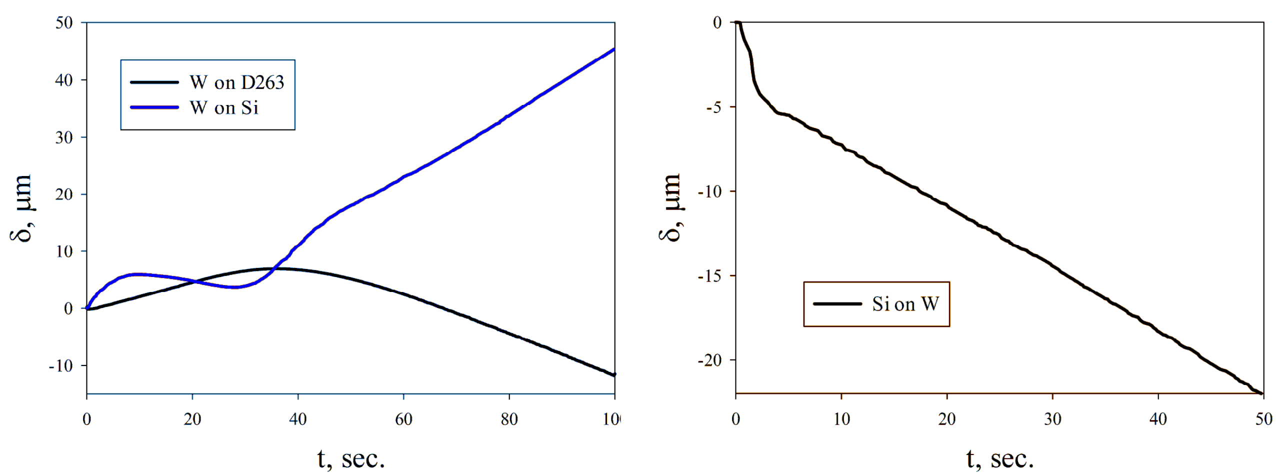

Figure 4. The cantilever tip displacement as a function of deposition time for the, W/glass, W/Si and Si/W interfaces. Unlike other high mass metals, W reaches a tensile steady-state when deposited on a sputtered layer of Si.

behavior and reaches a tensile steady-state when deposited on amorphous silicon, however. We have checked whether this effect also exists for other high density metals such iridium (Ir), but as shown in Fig. 5, the stress in the iridium remains compressive in the steady-state when deposited on a layer of amorphous Si. Evidently, the tungsten adatom mobility is uniquely influenced by the surface energy of the silicon layer, or alloying at the W/Si interface might occur, which somehow changes the microstructure of the tungsten layer. Continued work here is needed, however; to

understand more thoroughly the underlying mechanism for this behavior. In any case, the thickness of the constituent layers can be found which promote tensile stress in W/Si multilayers which can then be used to compensate the stress in W/Si x-ray optical coatings.

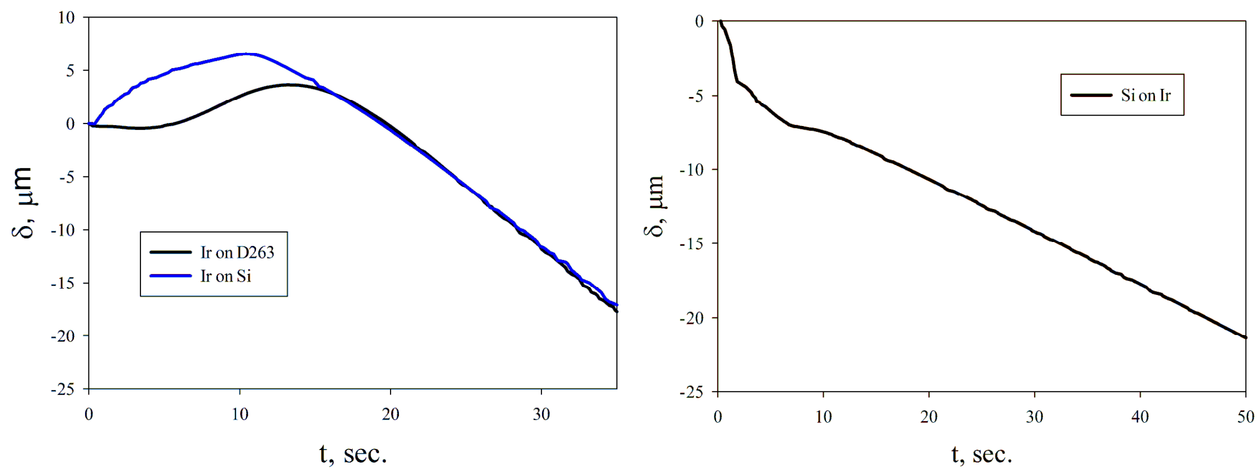

Figure 5. The cantilever tip displacement as a function of deposition time for the, Ir/glass, Ir/Si and Si/Ir interfaces.

2.2 W/Si stress compensating multilayer

The relevance of the in-situ stress measurement scheme demonstrated in §2.1 is to provide a quick assessment of whether a given set of deposition times can be found for a particular material pair to achieve a net tensile stress in the multilayer. The net tensile stress might then be used to compensate the compressive stress in x-ray or EUV optical coatings. The idea is illustrated in Fig. 6 for W/Si multilayers, but appropriate thicknesses can also be determined from Fig. 1 for Mo/Si multilayers as well. The thicknesses of the layers are chosen to give some net tensile stress in the multilayer, but are otherwise arbitrarily chosen in this example. The d-spacing of the compensating multilayer is ~7.3 nm with an interfacial roughness of ~0.45 nm. The layer thicknesses of the compensating multilayer might be optimized to maximize the tensile stress per bilayer while minimizing interfacial/surface roughness, for example. For illustrative purposes, we also show how the tensile stress is used to compensate the compressive stress in a multilayer composed of two different d-spacings. It is expected that any reasonable number of layers could be added to the compensating multilayer to balance arbitrarily large values of compressive integrated stress without appreciably impacting the performance of the optical coating, but more work is needed to confirm this. The compensating W/Si multilayer in this example can balance the compressive integrated stress in an optical coating with a magnitude of ~$6X10^4$ MPa-nm. This magnitude is sufficient enough to reduce the integrated stress by 75% in a W/Si optical coating that has an average stress of ~100 MPa of a total thickness of ~0.8 µm (i.e. ~$0.8X10^5$ MPa-nm). This value of compressive integrated stress is commensurate to that reported for the W/Si coatings used for the *NuStar* optics, for example[6].

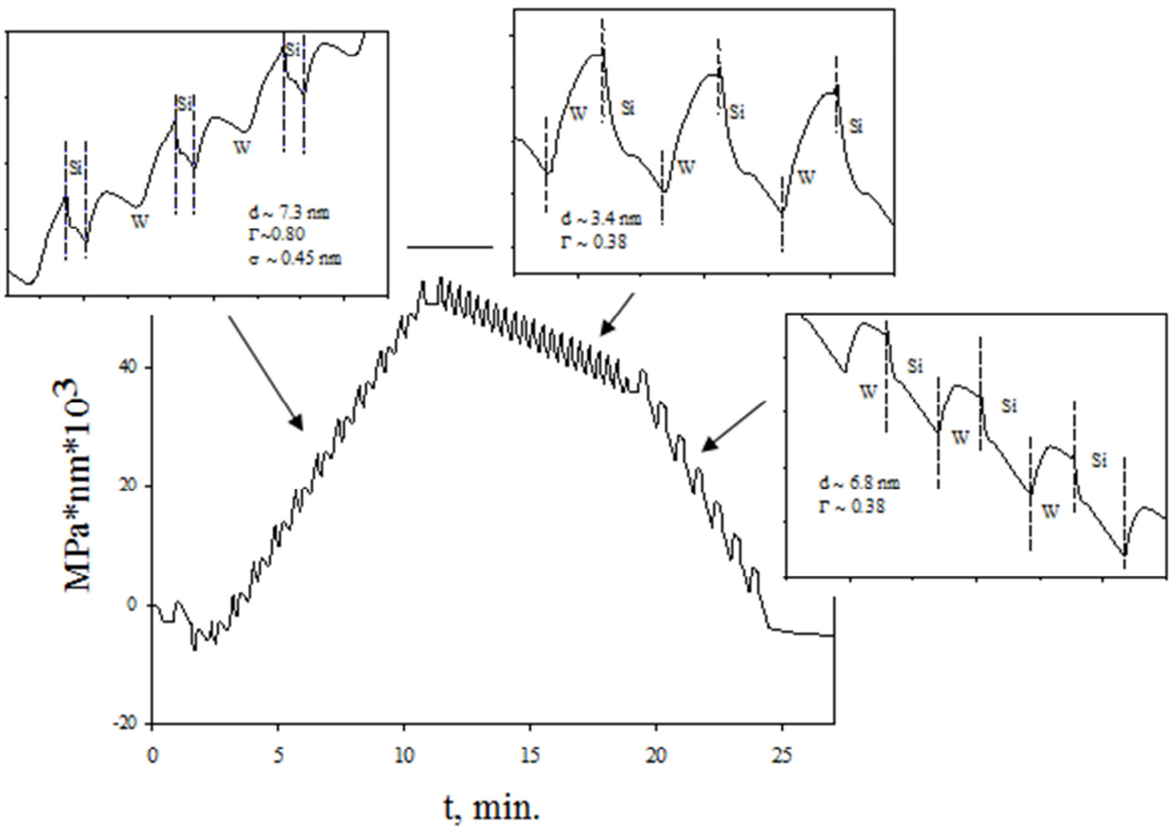

Figure 6. Illustration of how a W/Si multilayer in tensile stress can be used to balance the net compressive integrated stress in an x-ray optical coating. The lower Γ values (i.e. < 0.5) required in x-ray optical coatings often result in a net compressive integrated stress. The d-spacings of the optical coatings are arbitrarily chosen, and used here simply to demonstrate a concept.

2.3 Application to other material systems

For some material combinations, as we have checked, such as Ir/B_4C or W/B_4C, the metal layer will never become sufficiently tensile to allow their use for the practical application as a stress compensating multilayer. In this case a compensating multilayer composed of Mo/B_4C or Cr/B_4C might be used instead—requiring the use of a third material. The use of B_4C-based compensating multilayers may provide an additional advantage because several benefits have been observed when this material is reactively sputtered with nitrogen[4]. This includes a reduction in compressive stress of the B_4C layer, an increase in its deposition rate, and a reduction in interfacial roughness. For example, the use of nitrogen has been shown to significantly reduce the roughness of the Ir/B_4C interface[5]. The use of Ir/B_4C has appeal as an optical coating for future missions such as *Athena* and *Lynx*. The use of nitrogen might also be effective for further reducing the interfacial or surface roughness of other B_4C-based material combinations used as stress compensating multilayers like Cr/B_4C or Mo/B_4C.

3. CONCLUSIONS

We have introduced a method which utilizes multilayers, rather than a single layer film, to compensate the stress in x-ray and EUV optical coatings. This method could be advantageous because, unlike single layer films in tensile stress, the surface roughness may be less likely to increase with the total thickness of the multilayer. The use of in-situ stress measurement has been used to identify potential material combinations and the thicknesses of their constituent layers for achieving a net tensile integrated stress in the multilayer. A more detailed investigation into the influence of the stress compensating multilayer on the performance of the optical coating is needed and will be carried out in future work. This method is anticipated to add time to the deposition process, but may offer a reduction in complexity in comparison to other methods, particularly when the same material pairs are used for the compensating and optical coating.

Optimization of the layer thicknesses to minimize the number of bi-layers within the constraints of low surface roughness is also needed to more accurately assess the impact to the deposition time.

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